Ultrafast Probes for Dirac Materials

Dmitry Yarotski

Center for Integrated Nanotechnologies
Materials Physics and Applications Division
Los Alamos National Laboratory

Quantum and Dirac Materials Workshop
March 8-11, 2015, Santa Fe, NM, USA

Los Alamos
NATIONAL LABORATORY
EST. 1943

Collaborators and Acknowledgements

LANL Staff: Rohit Prasankumar, Antoinette Taylor, Abul Azad, Steve Gilbertson, George Rodriguez, Tomasz Durakiewicz, Aditya Mohite, Andrew Dattelbaum, Quanxi Jia, Stuart Trugman, Jian-xin Zhu

LANL Postdocs: Rolando Valdes Aguilar, Yaomin Dai, Keshav Dani, John Bowlan, Jingbo Qi, Jinho Lee, Georgi Dakovski

Brookhaven National Laboratory: Genda Gu, Ruidan Zhong

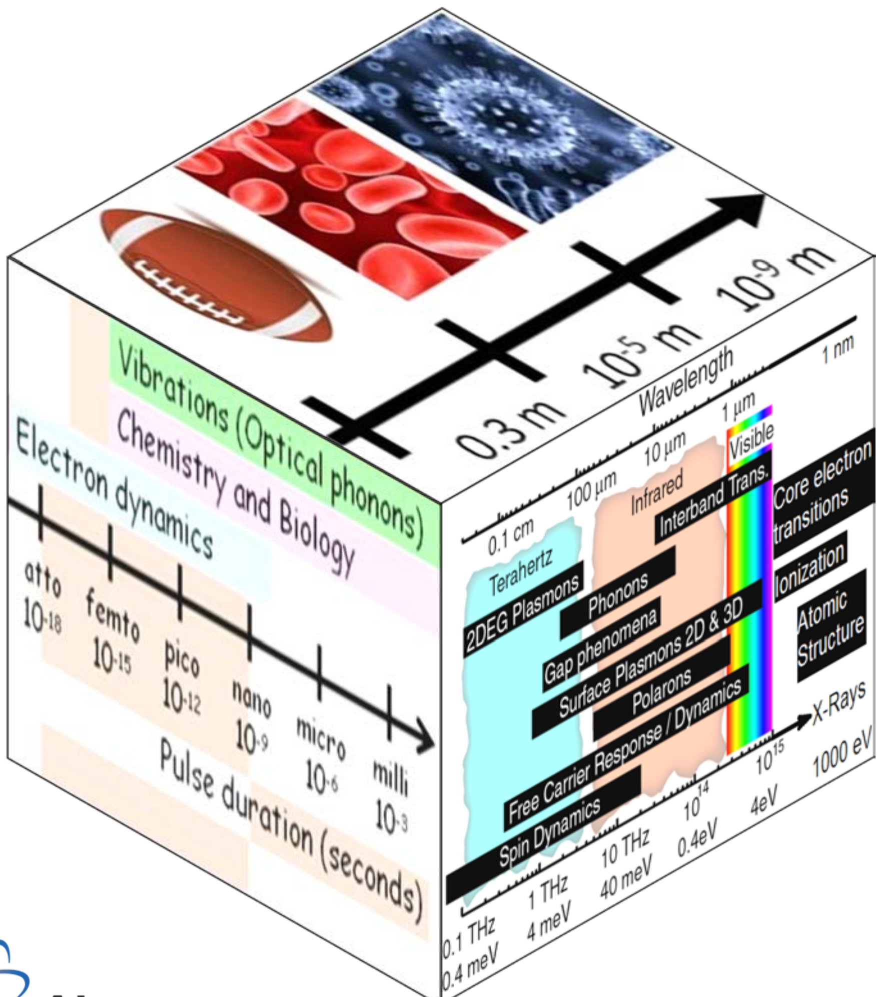
Rutgers University: Matthew Brahlek, Namrata Bansal, Seongshik Oh

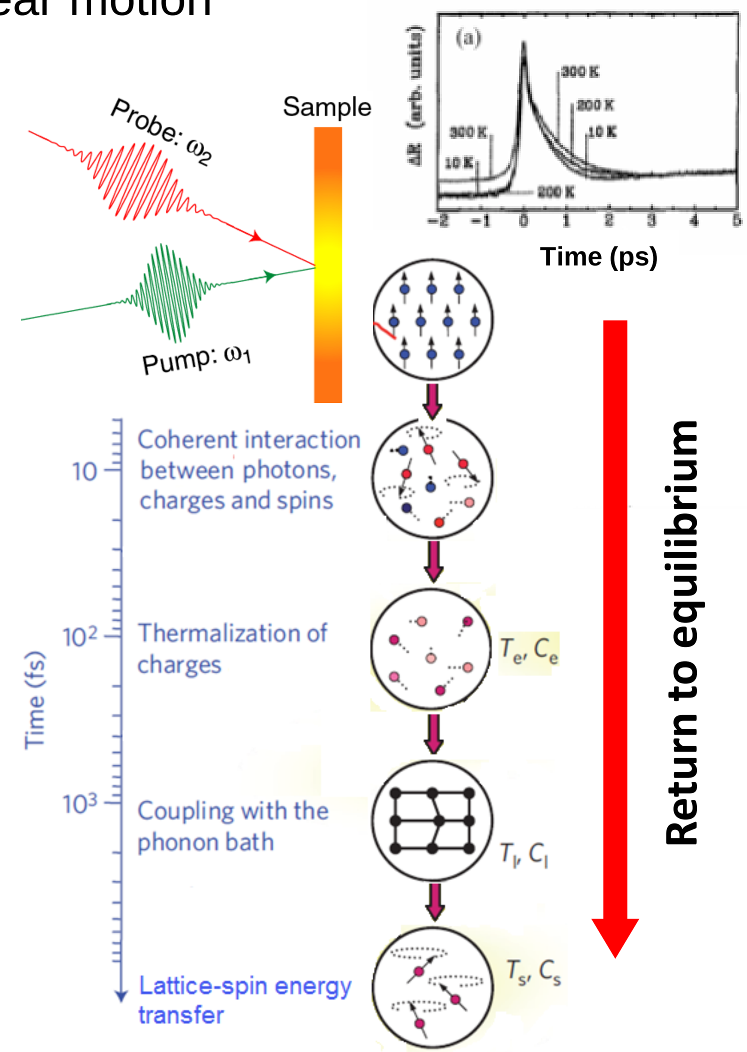
Rice University: Sina Najmaei, Jun Lou, Pulickel M. Ajayan,

We gratefully acknowledge the support from the U.S. Department of Energy through the Center for Integrated Nanotechnologies, LANL LDRD Program, and the UC Office of the President under UC Lab Fees Research Program

Why Ultrafast Spectroscopy ?

Ultrafast (10-100 fs) spectroscopy can resolve non-equilibrium dynamics (quasiparticle, transport etc.) at the fundamental time and spatial scales of electronic and nuclear motion

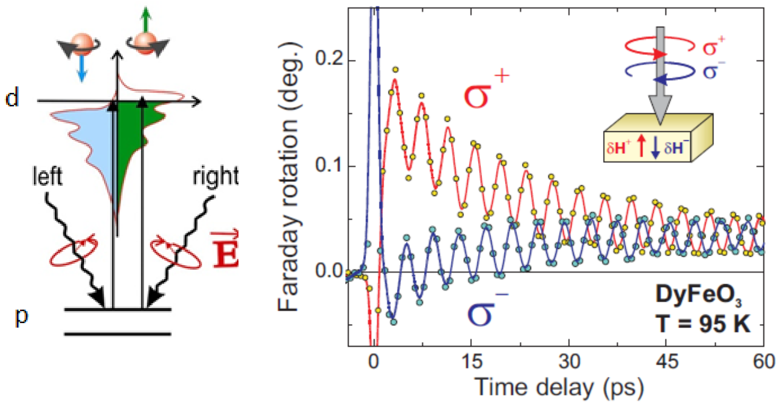




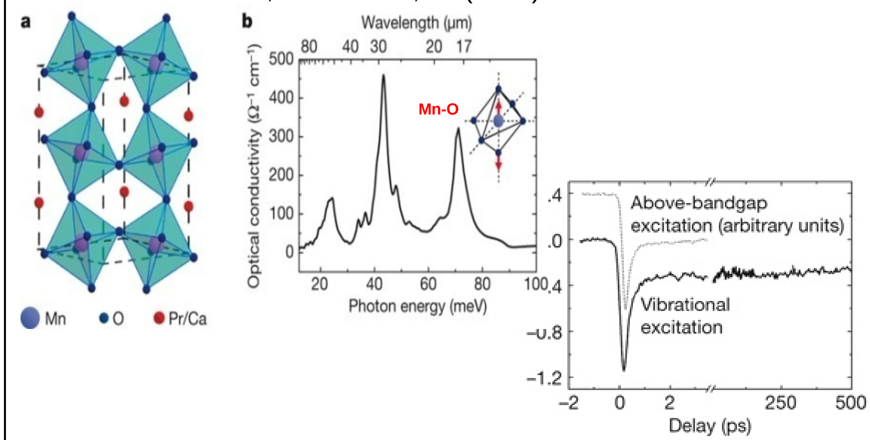
Ultrafast Coherent Order Manipulation

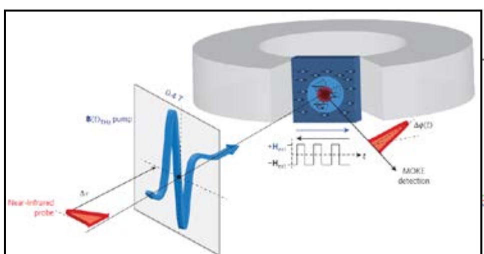
Manipulation of order parameters ◆ Photoinduced phase transitions ◆ New non-thermally accessible phases.

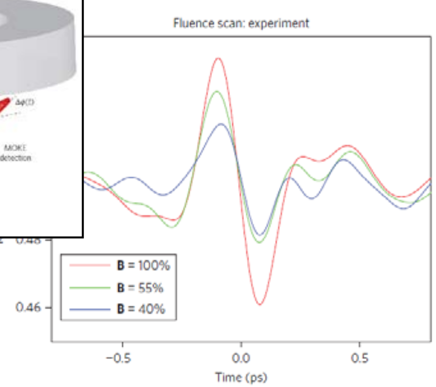
Kimel et al., Nature 435, 655 (2005)



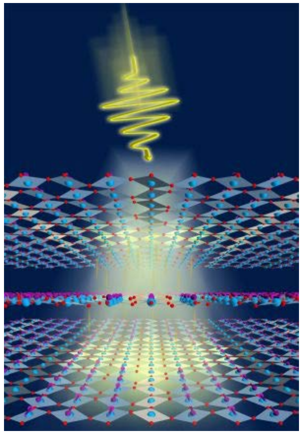
Rini, Nature 449, 72 (2007)

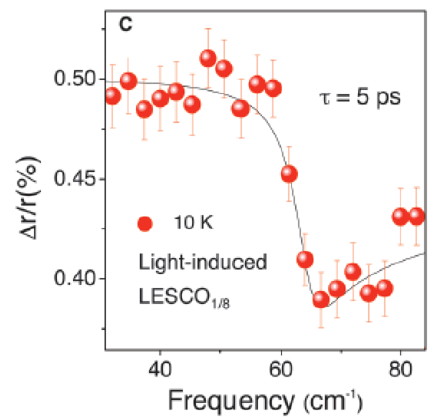






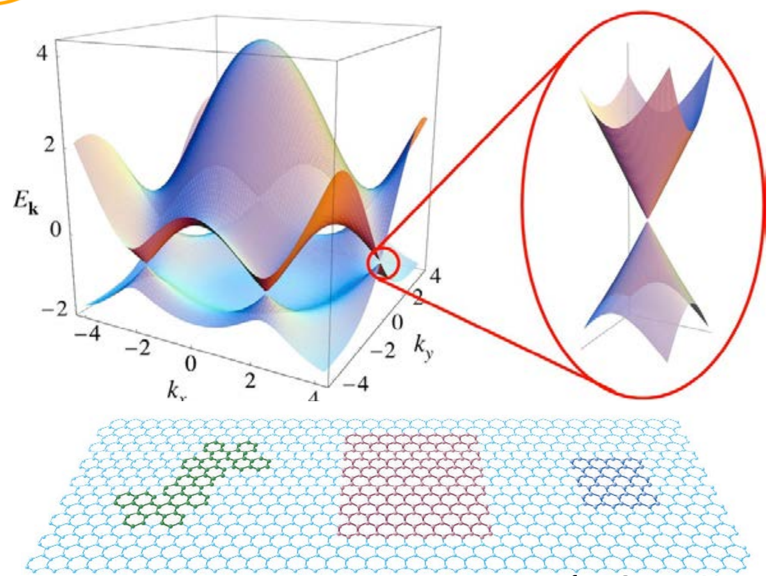
Vicario, Nature Phot. 2013





Fausti, Science 2011

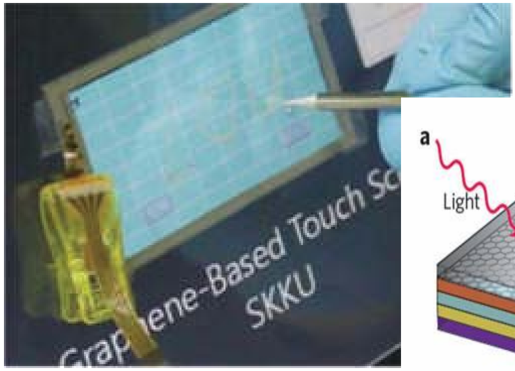
Graphene: The Slice that Started It All



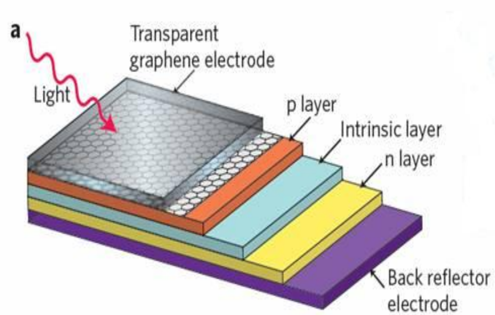
after Castro Neto

- **<u>Graphene:</u>** a basis for 0D buckyballs, 1D carbon nanotubes, and 3D graphite
- Quasiparticles are described by relativistic Dirac equation – ***Dirac Material***
- Massless Dirac quasiparticles exhibit novel transport properties (high mobility, excellent conductivity)

Understanding the *non-equilibrium behavior of photoexcited graphene* is important for science and applications in detectors, solar cells and displays.



Bae et al. *Nat. Nanotech*. 2010



Bonaccorso et al. *Nat. Photonics* 2010

Quasiparticles in Graphene

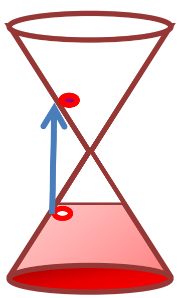
Linear dispersion near Dirac point gives for relativistic quasiparticles:

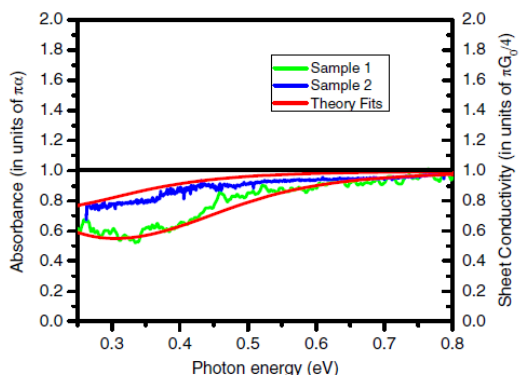
$$E \approx \hbar v_F k \qquad\qquad E_F^{e,h} \sim \hbar v_F \sqrt{\pi N_{e,h}}$$

Are *photoexcited* quasiparticles in graphene relativistic too?

Two types of optical conductivity in graphene:

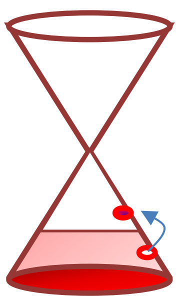
Interband is *constant* in a wide spectral range (flat 2.3% absorption)

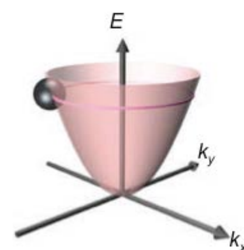




Mak et al., Phys. Rev. Lett. (2008)

Intraband differs for linear and parabolic bands





$$\sigma_{intra}^{mass} = \frac{Ne^2}{m}\frac{\gamma}{\omega^2+\gamma^2}$$

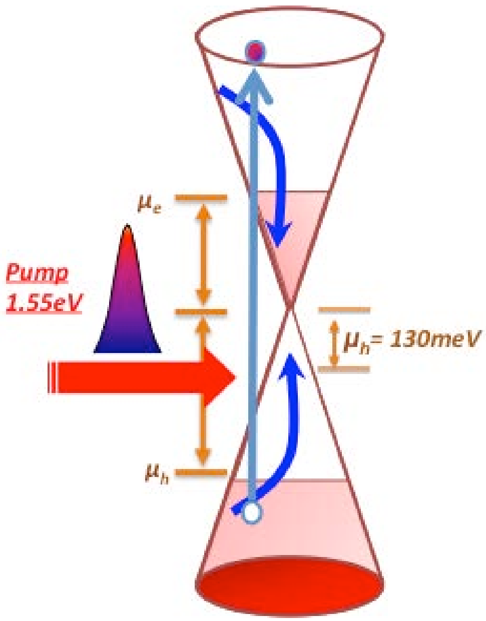
$$\sigma_{intra} \propto \sqrt{N}\frac{\gamma}{\omega^2+\gamma^2}$$

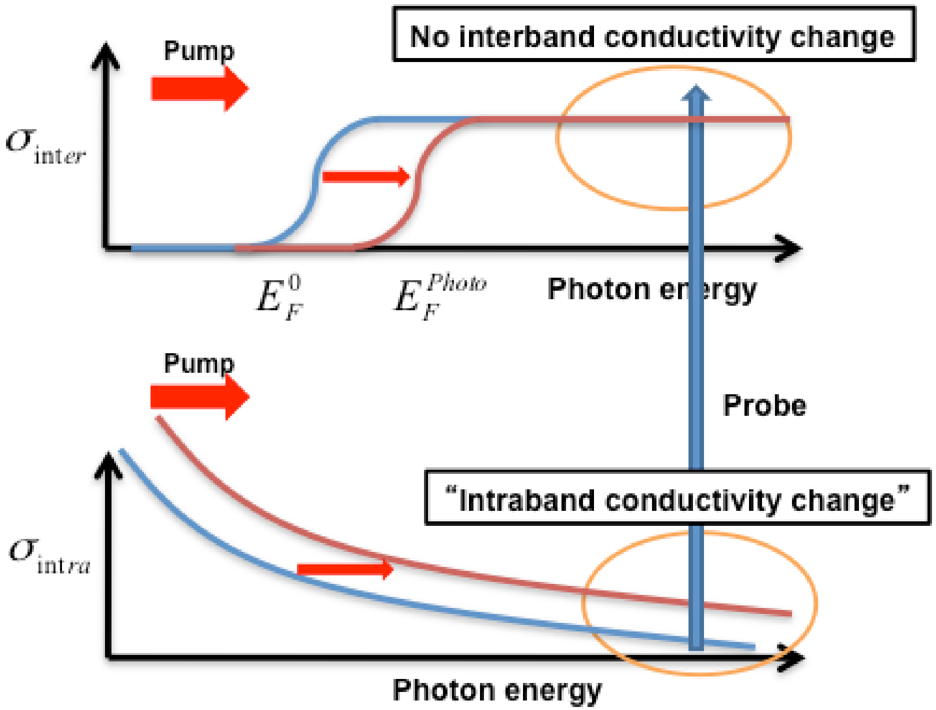
Measuring conductivity change after photoexcitation as function of *N* will indicate whether non-equilibrium quasiparticles are relativistic

Measuring Relativistic Quasiparticles in Graphene

We measure the photoinduced conductivity change:

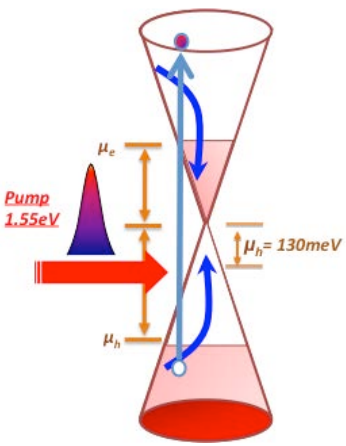
$$\Delta\sigma = (\sigma_{inter} + \sigma_{intra})\big|_{Photo-excited} - (\sigma_{inter} + \sigma_{intra})\big|_{Intrinsic\ \ doping}$$

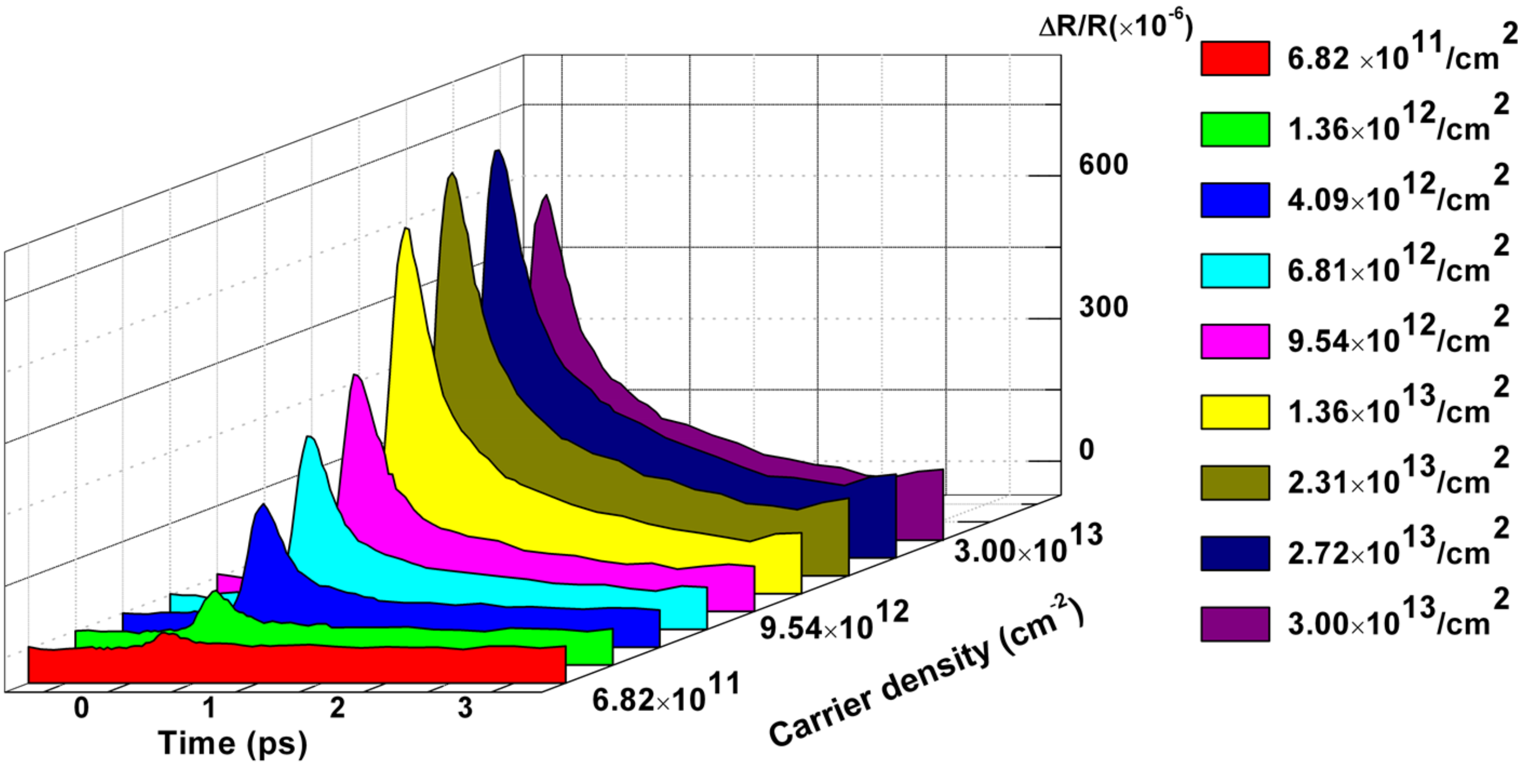




The change in conductivity, as measured in a visible pump-probe experiment, is dominated by the intraband component!

Near-IR Pump, Visible-Probe Spectroscopy



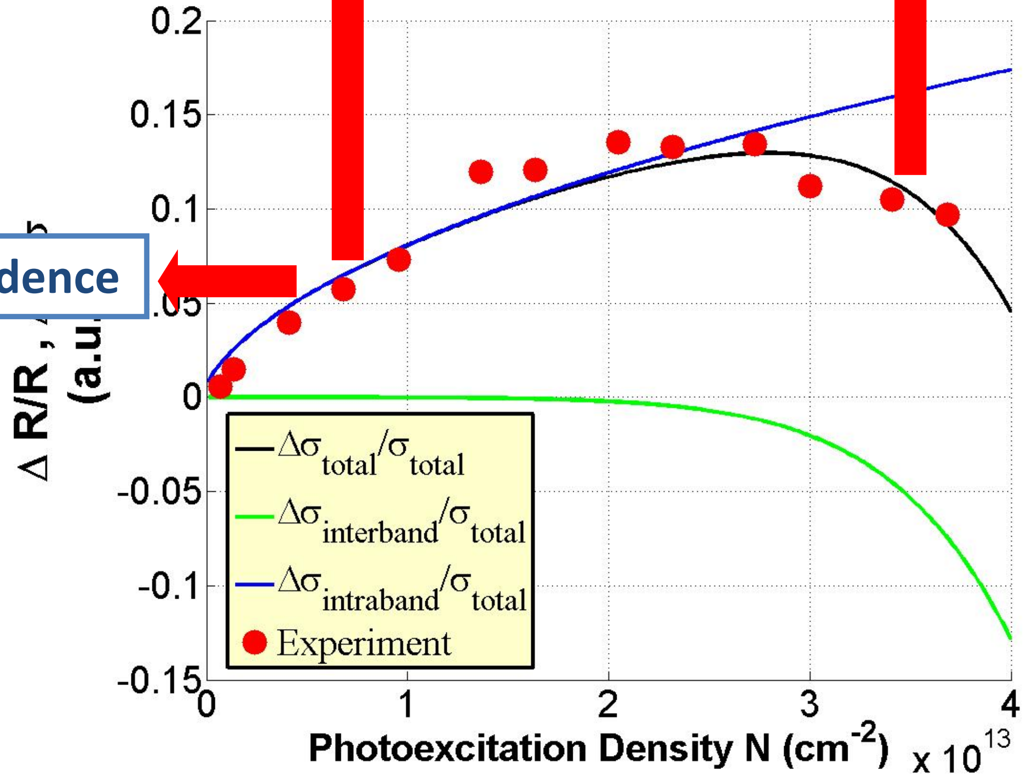


- 1.55 eV pump, 1.77 eV probe experiments
- Fermi energy after photoexcitation = 700 meV (for N~3.1x10^{13}/cm^2)
- Decay dynamics are qualitatively identical for all photon energies (1.74-2.42 eV)
- Electron-electron thermalization within <100 fs – **Amplitude gives optical Δσ**
- Electron-phonon thermalization within 1.4 ps

Hot Dirac Fermions in Graphene

Intraband contribution | **Inter**band contribution

$\propto \sqrt{N_{e,h}}$ **dependence**



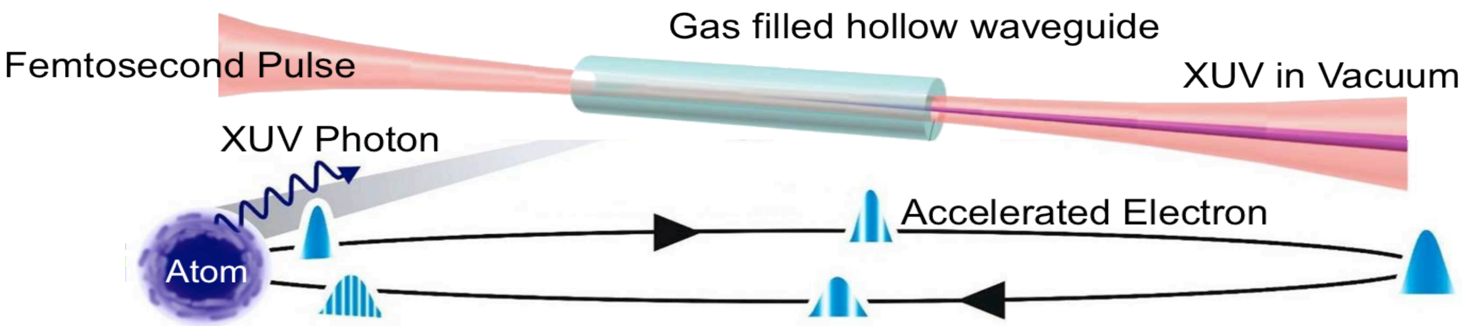
Reflectivity (or conductivity) change follows $\sqrt{N}$ from $E_F^{e,h} \sim \hbar v_F \sqrt{\pi N_{e,h}}$

Our experiment reveals the relativistic nature of photoexcited Dirac quasiparticles in graphene

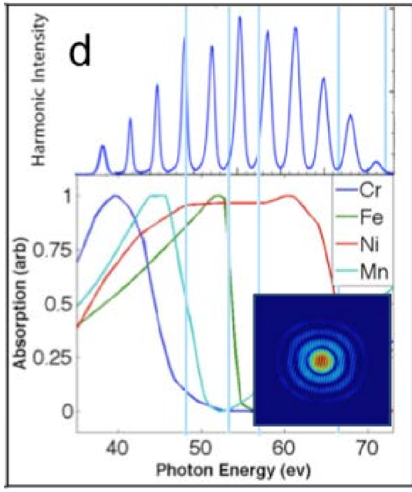
K. M. Dani et al, *Phys. Rev. B* (2012)

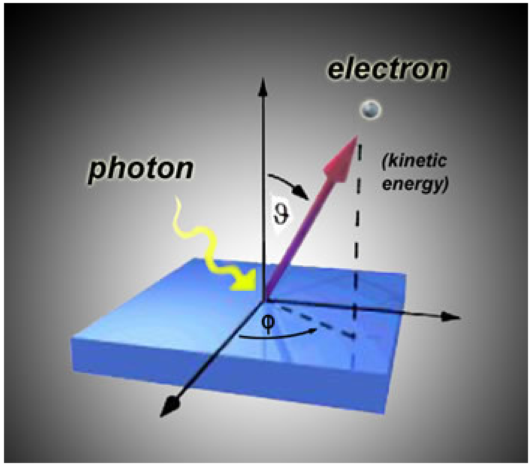
Time-Resolved ARPES

High Harmonic Generation – Extreme nonlinear frequency upconversion



M. Ferray, *et al.* J. Phys., 21 (1988); P.B. Corkum, PRL **71**, 1994 (1993)



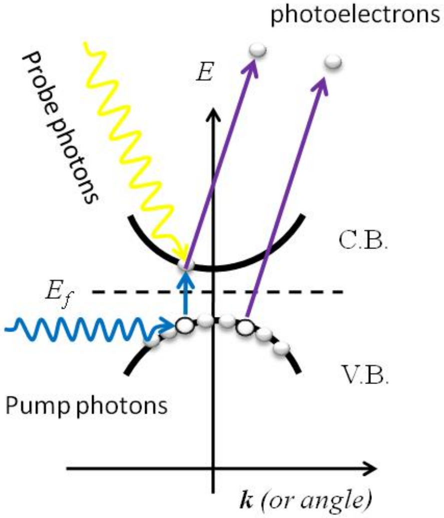


STATIC ARPES:

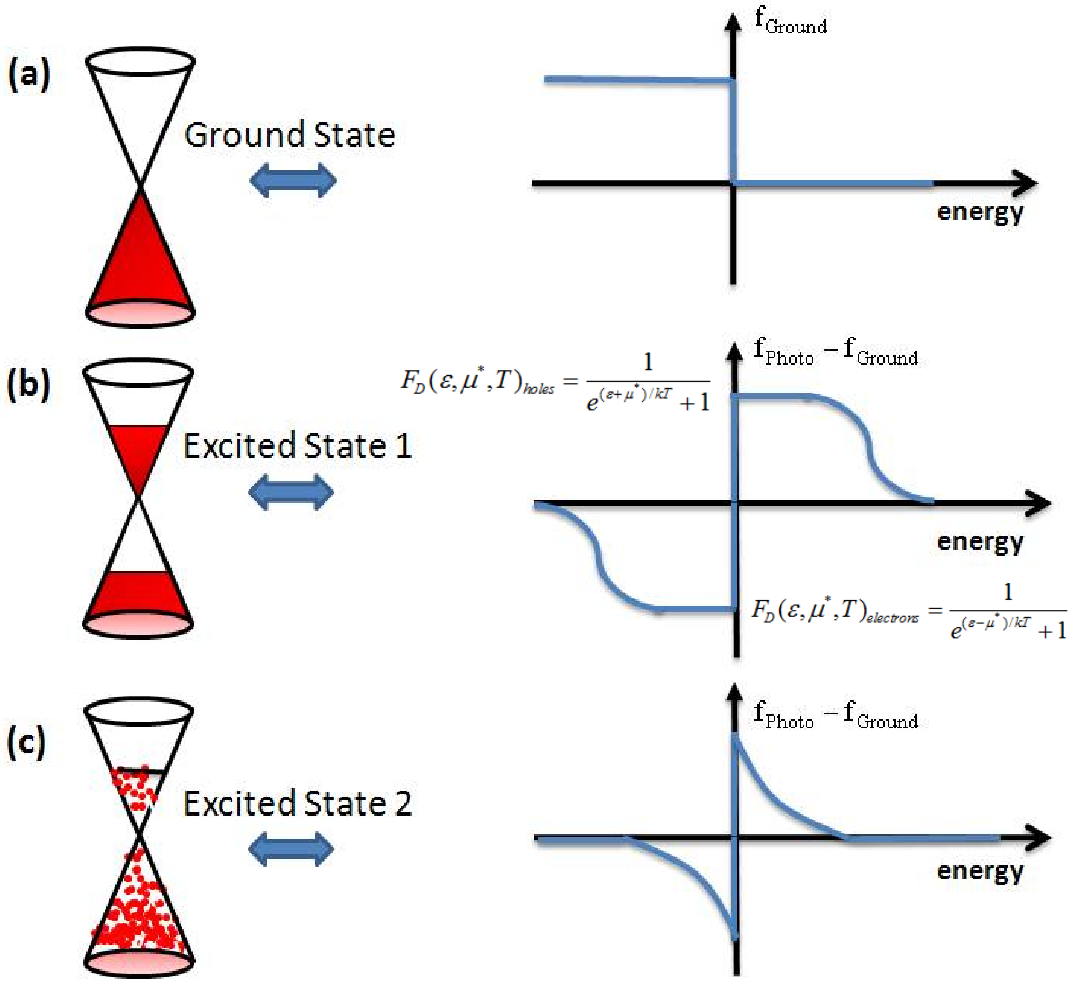
- probes electronic structure in both **E** and ***k*** domains

DYNAMIC ARPES:

- probes transient electronic structure changes in both **E** and ***k*** domains
- Fills excited states to reveal their structure



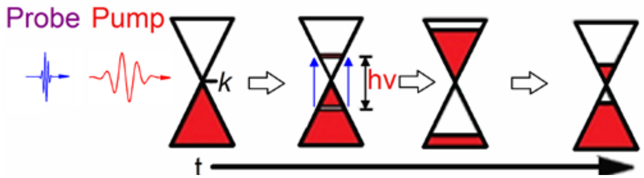
Photoexcited Fermi-Dirac Distribution in Graphene

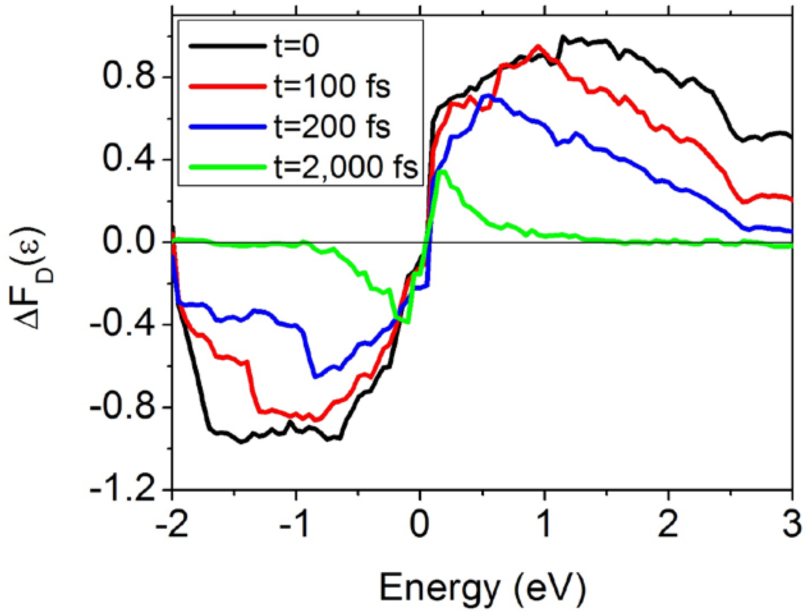


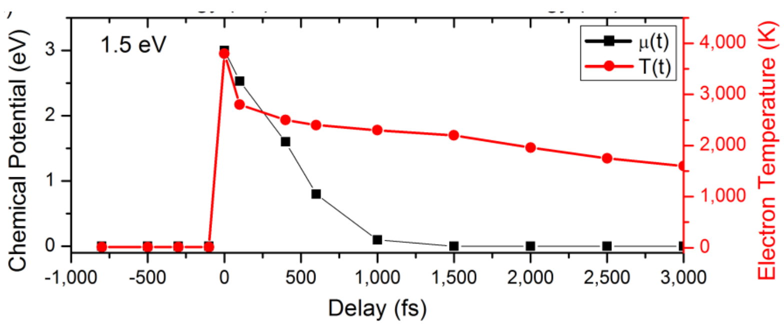
$$F_D(\varepsilon,\mu,T^*) = \frac{1}{e^{(\varepsilon-\mu)/k_B T^*}+1}$$

- Is the Fermi-Dirac distribution of photoexcited carriers in graphene more like a metal (same μ_e and μ_h) or like a semiconductor (separate μ_e and μ_h)?
- Do processes like Auger recombination influence the dynamics at early times?
- Time-resolved photoemission experiments show that, in our samples, the photoexcited carriers retain separate F-D distributions for a few hundred femtoseconds

Recombination of Electronic States in Graphene



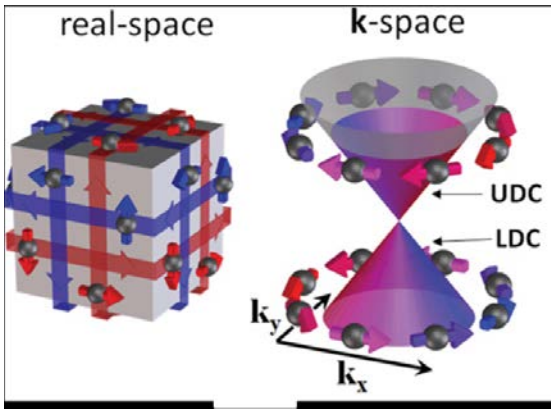




- ❖ Ultrafast pump/probe experiment on CVD grown graphene
  - 30 fs IR pump and sub-10 fs, 30-eV probe via HHG
  - measure tr-ARPES
- ❖ A short-lived distribution of carriers and holes is formed after optical excitation.
- ❖ Separate populations are:
  - ◆ semi-conductor like ($\mu^* \neq 0$) at early delays
  - ◆ metallic like ($T^* \neq 0$) at later times

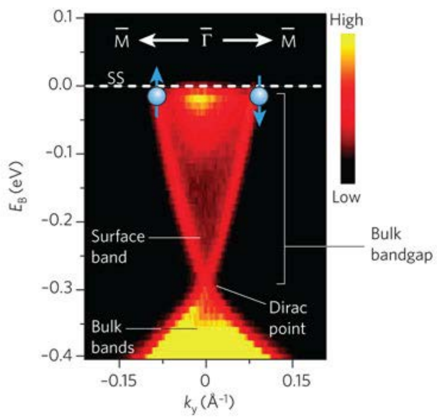
S. Gilbertson et al,, JCP Letters (2012)

Topological Insulators



* after A. Lanzara

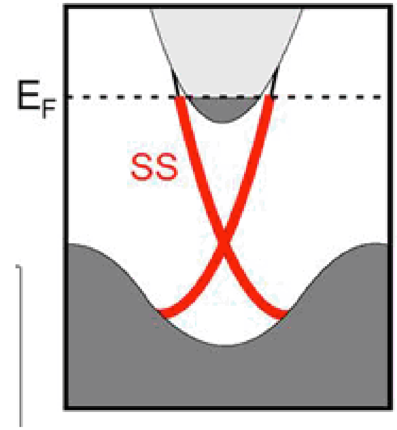
Moore et al., Nature 2010



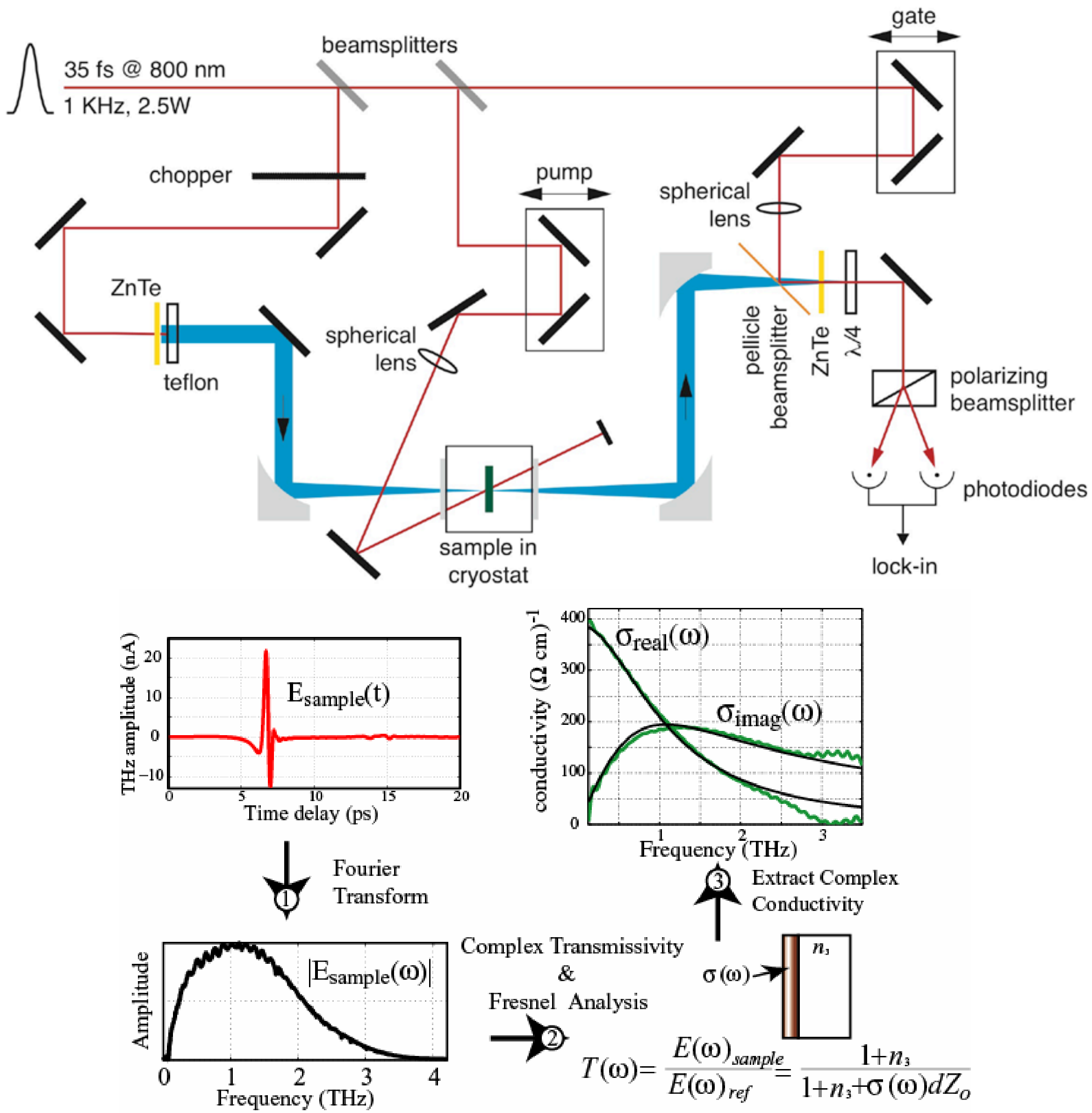
<u>Materials with exotic surface states</u>

- Linear *E-k* dispersion
- TRS protection against scattering
- Locked spin-***k*** relationship
- Majorana Fermions
- Spintronics, optoelectronics

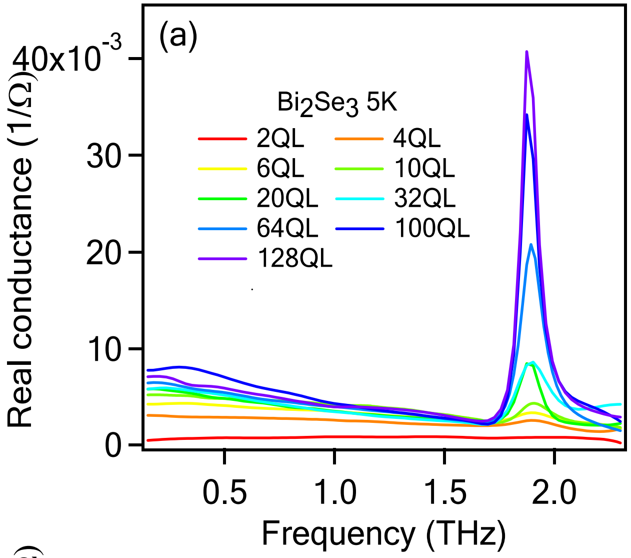
- **Real materials are not ideal – dopants/defects result in significant bulk interference**
- **THz spectroscopy provides the ability to separate the collective motion of charge carriers in bulk vs. surface states**

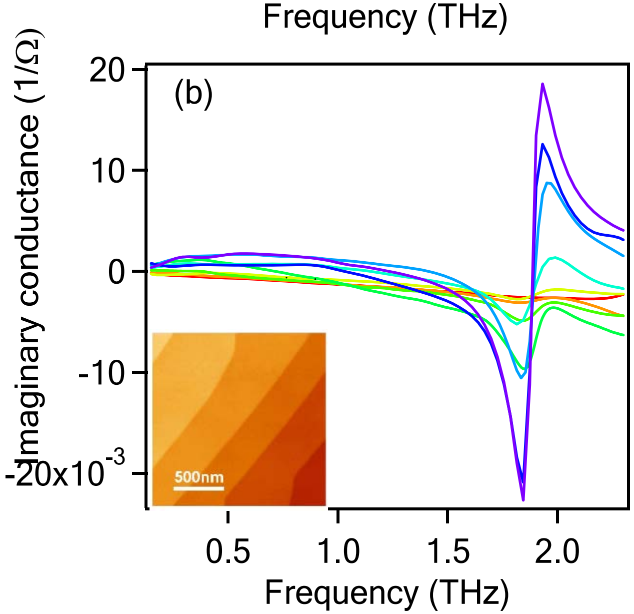


Optical Pump Terahertz Probe

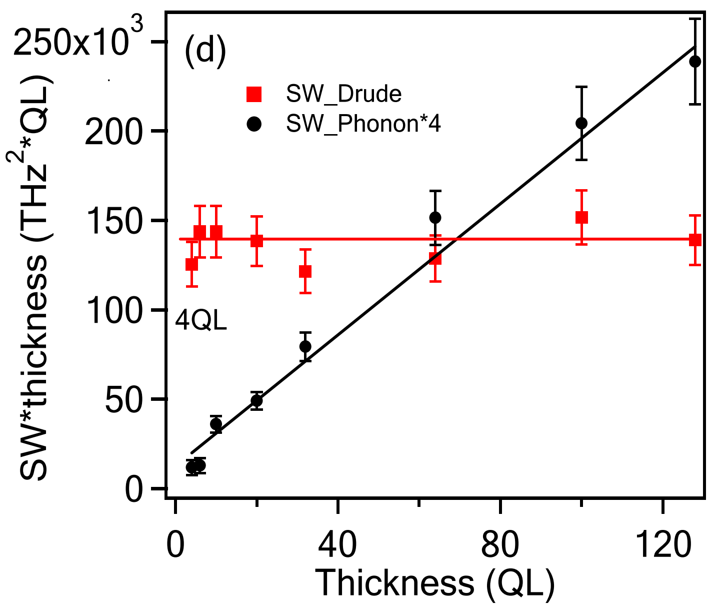


Terahertz Conductivity of Bi_2Se_3





- **Low freq. spectra:**
  Drude component: $1/\tau \sim 1$ THz
  Bulk phonon: $\omega_0 \sim 1.9$ THz
- **Electron density consistent with**
  $n_{surf} \sim 1.5 \times 10^{13}$ cm^{-2}
- **Drude term is thickness independent**
  Surface.
- **Phonon is not → Bulk effect.**



Time-Resolved THz Spectroscopy

Fix THz gate delay at maximum and scan pump-probe delay

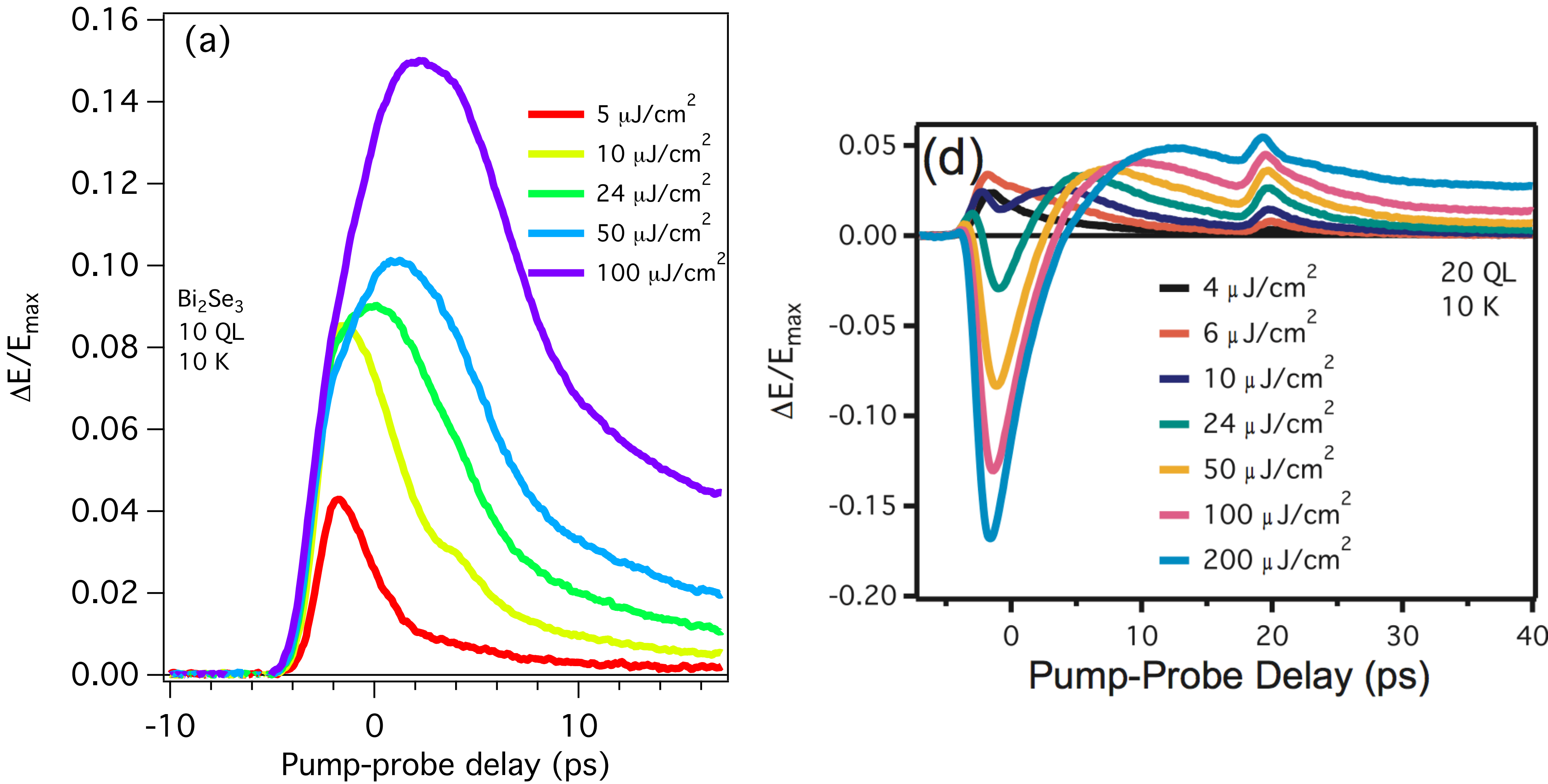
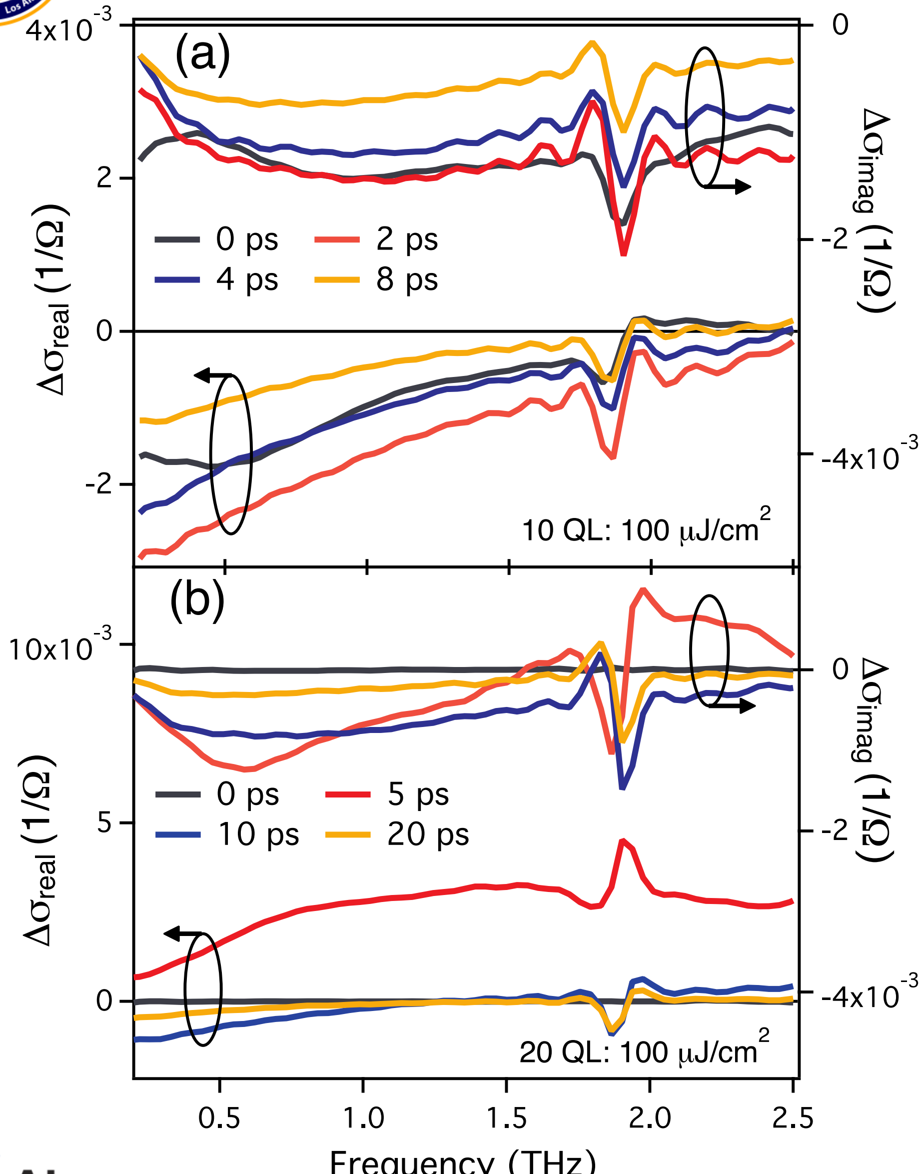


Photo-Induced Conductivity in Bi_2Se_3



- **Drude-Lorentz Model:**

$$G_j(\omega) = \left(-\frac{\omega_{pD,j}^2}{i\omega - \Gamma_{\mathrm{Drude},j}} - \frac{i\omega\omega_{pDL,j}^2}{\omega_{DL,j}^2 - \omega^2 - i\omega\Gamma_{\mathrm{Lorentz},j}} - i\left(\varepsilon_\infty - 1\right)\omega \right) \varepsilon_0 d$$

- **Well described by *single* carrier type**
- **Carriers in 20 QL decay faster**

- **Green**: Drude (free electron).
- **Purple**: Phonon.

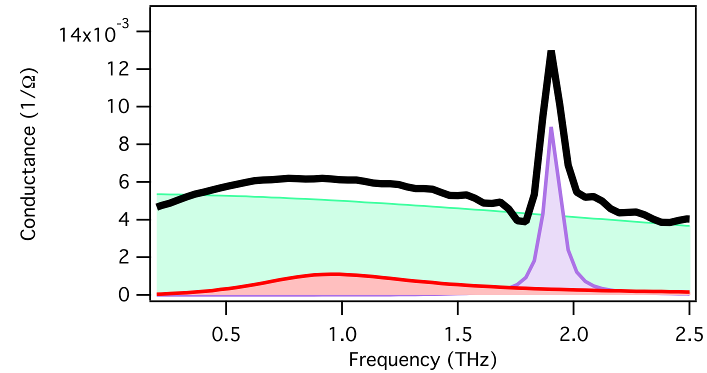
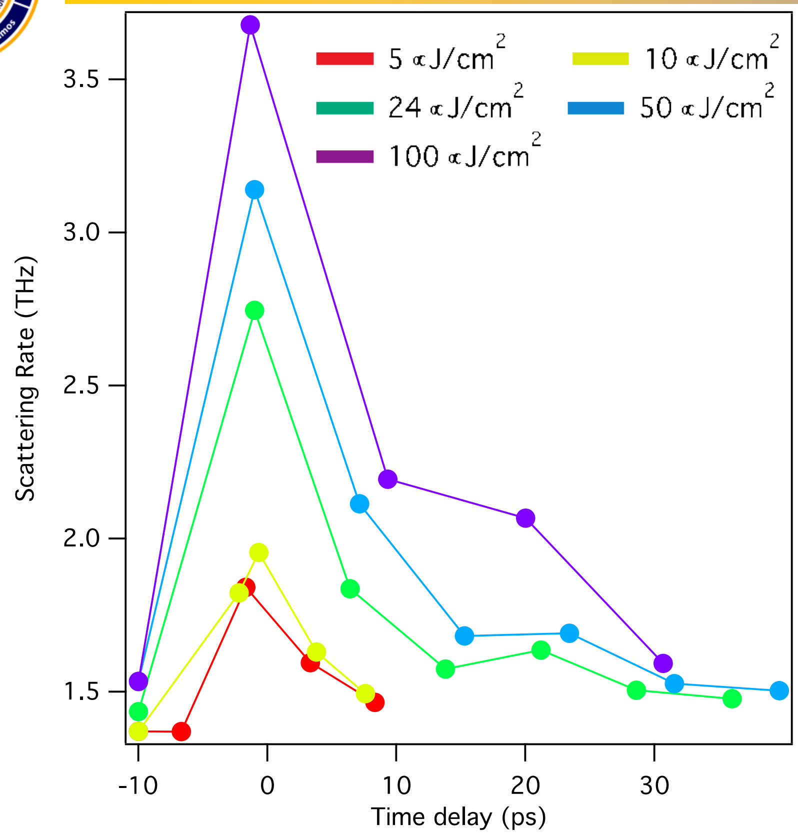
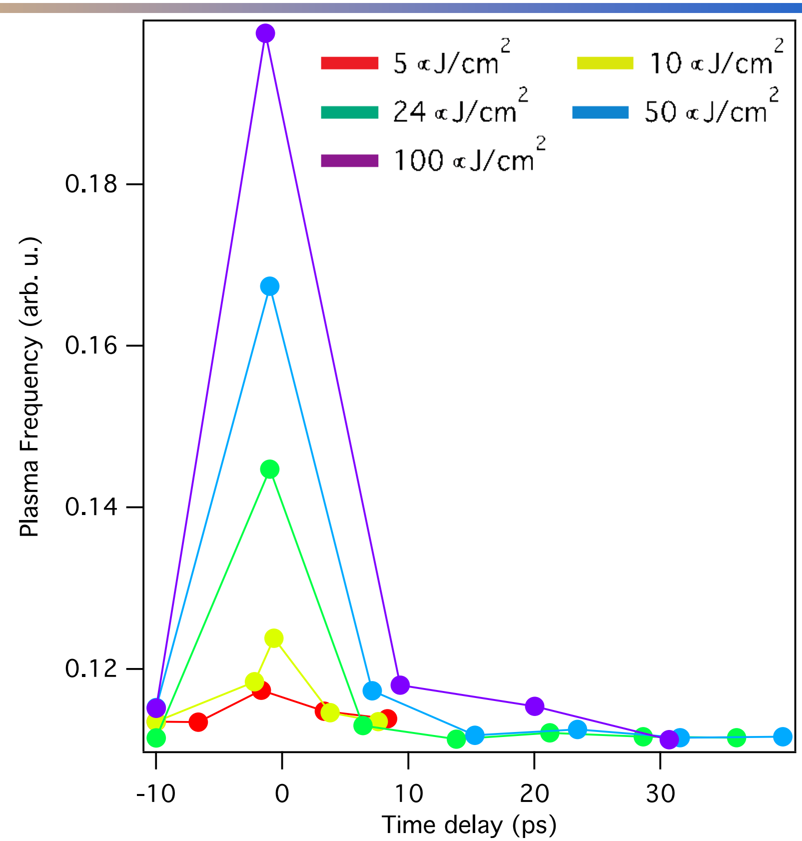


Photo-Induced Drude Properties in 20 QL





Low Fluence: increase scat. rate -> increase T

High Fluence: increase plasma freq. -> decrease T

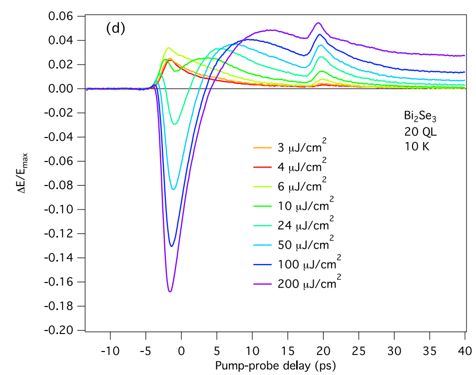
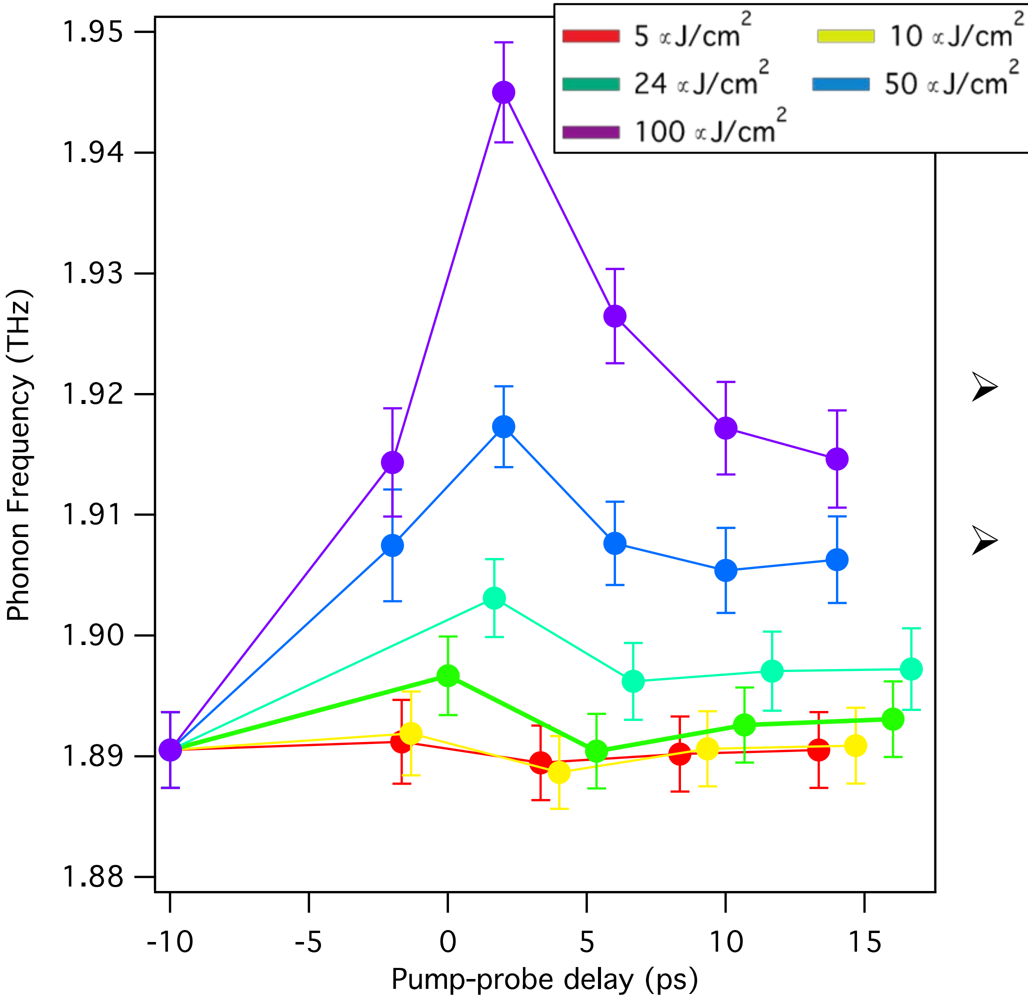
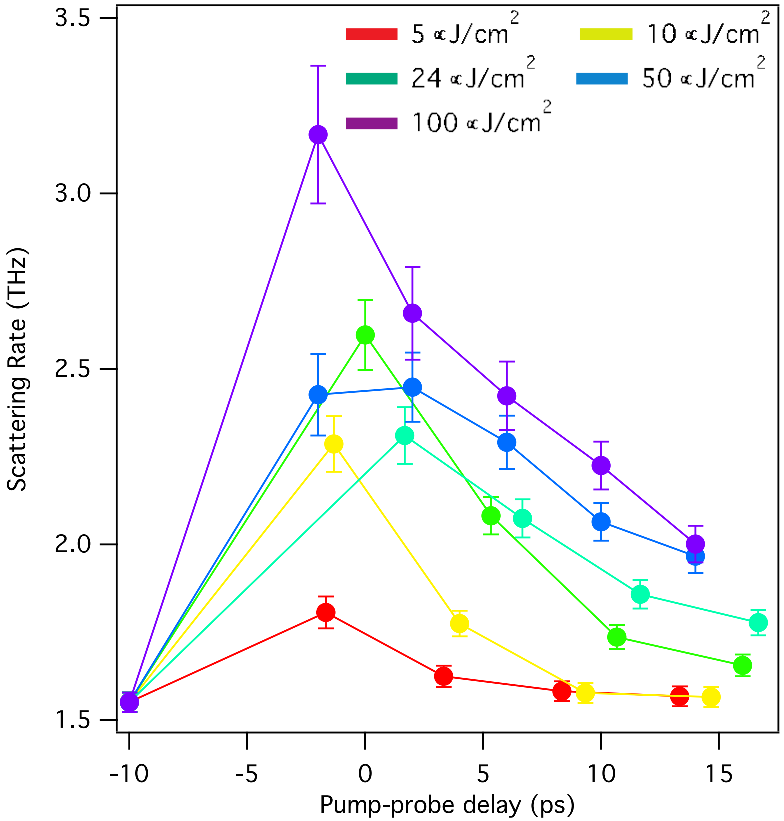


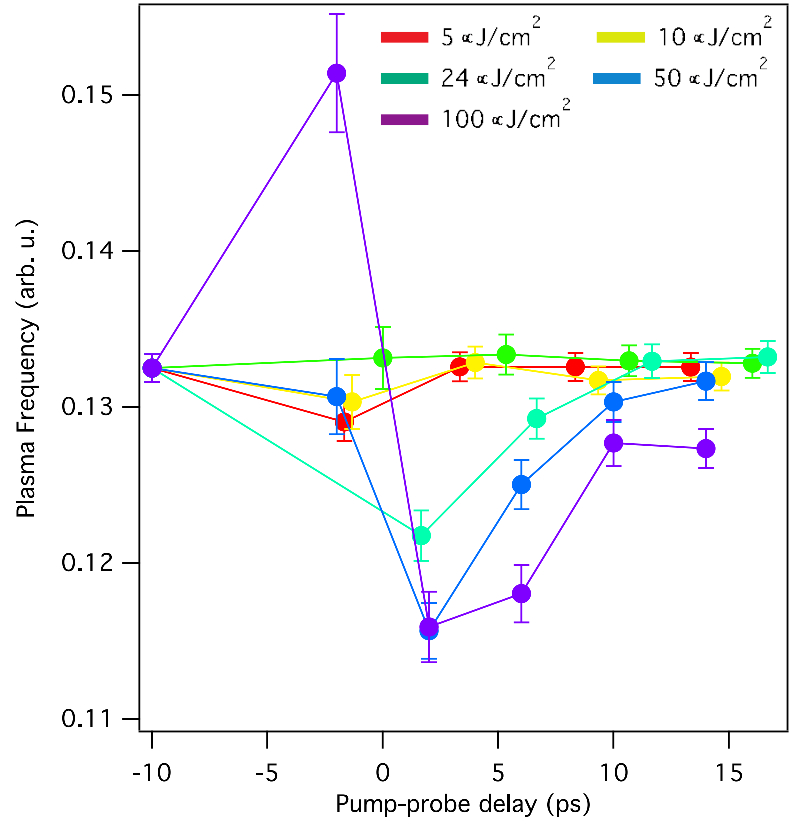
Photo-Induced Phonon Frequency Shift in 20 QL



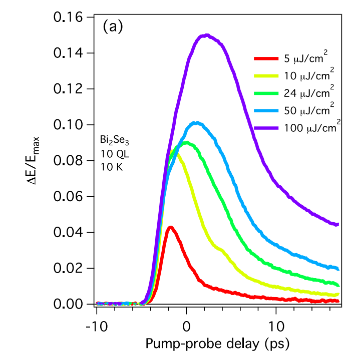
- **At high fluence, phonon shifts - similar to increase in temperature.**
- **Highest lattice temperature ~ 200 K**

Photo-Induced Drude Properties in 10 QL





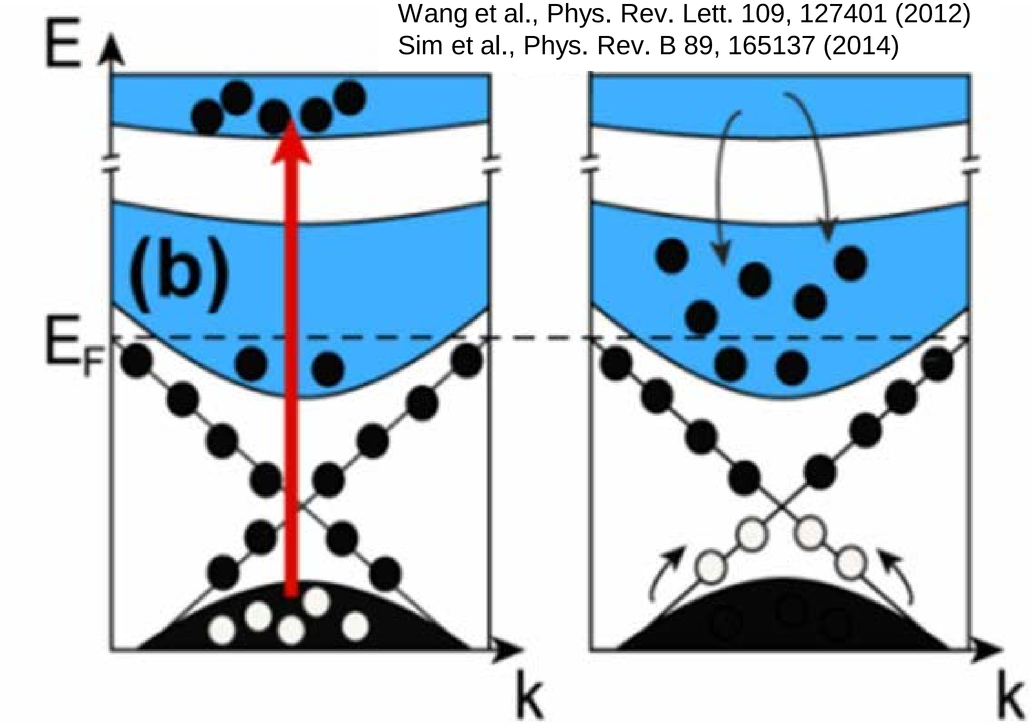
- Plasma frequency doesn’t change as much as in 20 QL sample.
- Scattering rate does, so the sample becomes more transparent at higher fluence.



Physical Picture

Phonon-induced bulk-to-surface scattering is not effective below T_D=180 K

Wang et al., Phys. Rev. Lett. 109, 127401 (2012)
Sim et al., Phys. Rev. B 89, 165137 (2014)



Hot surface carriers can be accessed independently from the bulk ones using THz spectroscopy

Thin 10 QL films are similar to graphene:

- Surface electrons dominate, but $\Delta\omega_p$ is small
- Γ_{surf} increases due to e-h scattering and temperature rise (~200 K) due to e-ph relaxation

Thick 20 QL films:

- Surface response dominates at low fluences
- High fluences result in large number of bulk carriers => higher $\Delta\omega_p$ and Γ_{bulk}
- Bulk electrons decay in ~5 ps
- Surface electrons decay in 20 ps preserving high scattering rates

Topological Crystalline Insulators

PRL **106,** 106802 (2011)   PHYSICAL REVIEW LETTERS   week ending 11 MARCH 2011

Topological Crystalline Insulators

Liang Fu

Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA

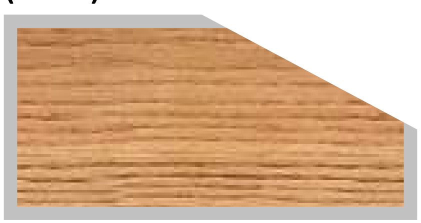
(Received 5 October 2010; revised manuscript received 31 December 2010; published 8 March 2011)

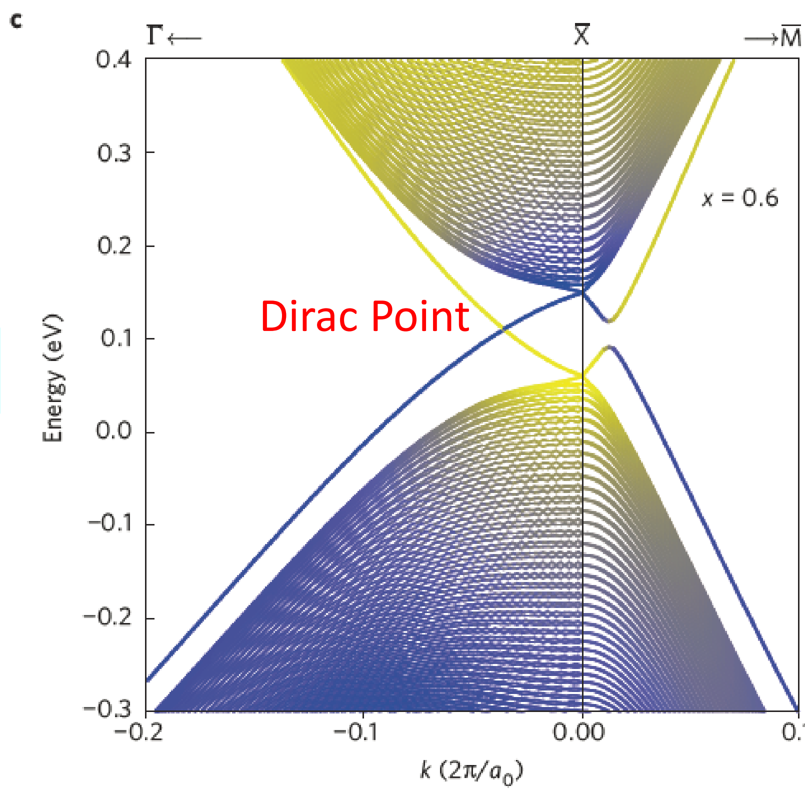
TI → Time Reversal Symmetry

TCI → Crystalline Symmetry

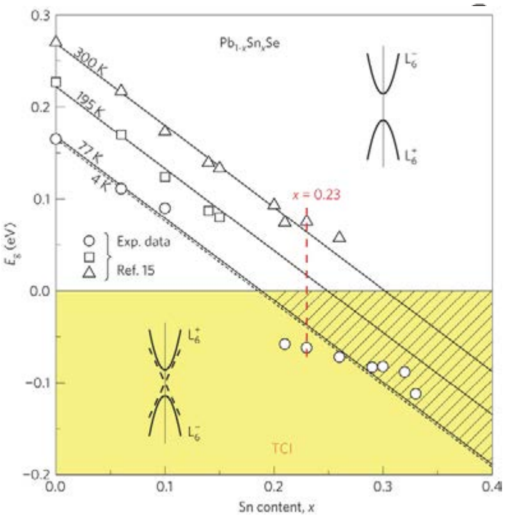
Metallic states on High Symmetry surfaces!

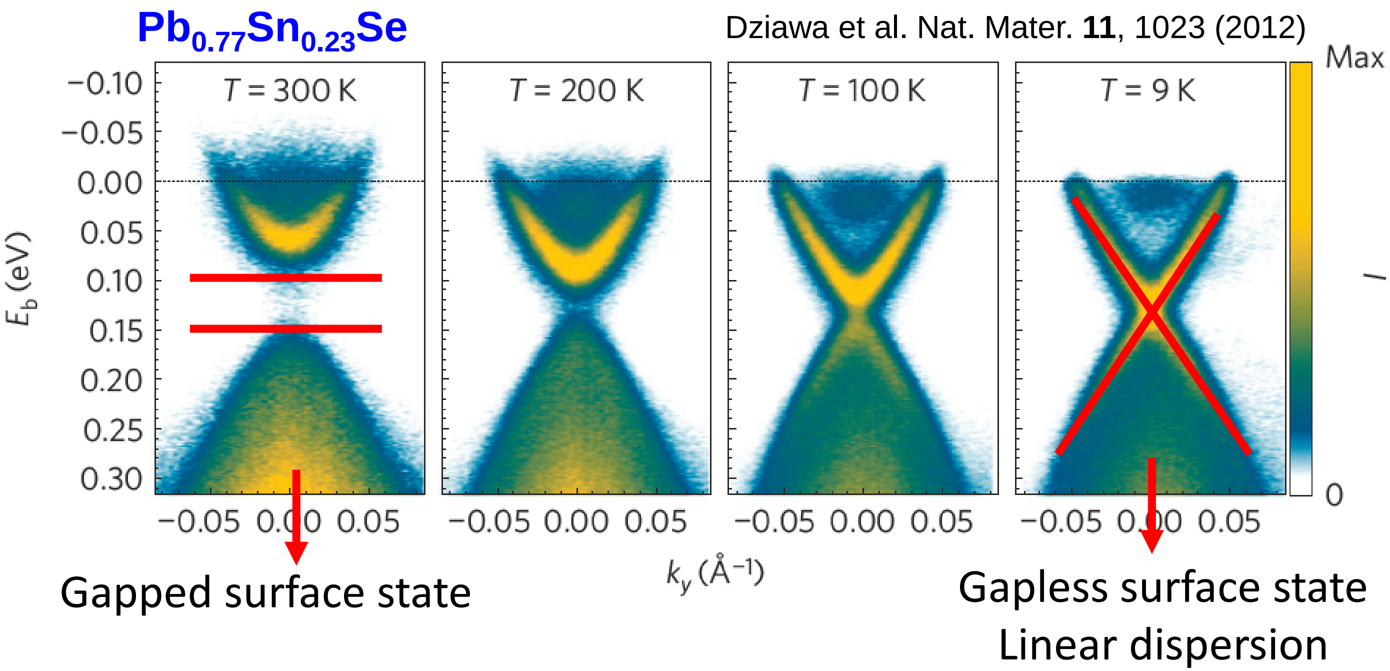
(001) surface





Topological Phase Transition in $Pb_{1-x}Sn_xSe$





P-induced TPT in $Pb_{1-x}Sn_xSe$ Xi et al. PRL **113**, 096401 (2014)

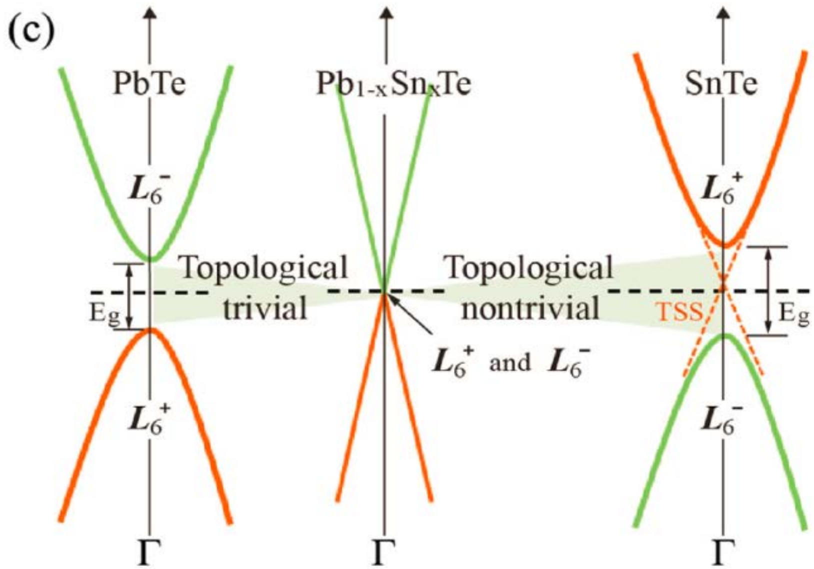
Topological Phase Transition in $Pb_{1-x}Sn_xTe$

PbTe
Trivial

Doping-driven Topological phase transition

$Pb_{1-x}Sn_xTe$

SnTe
TCI



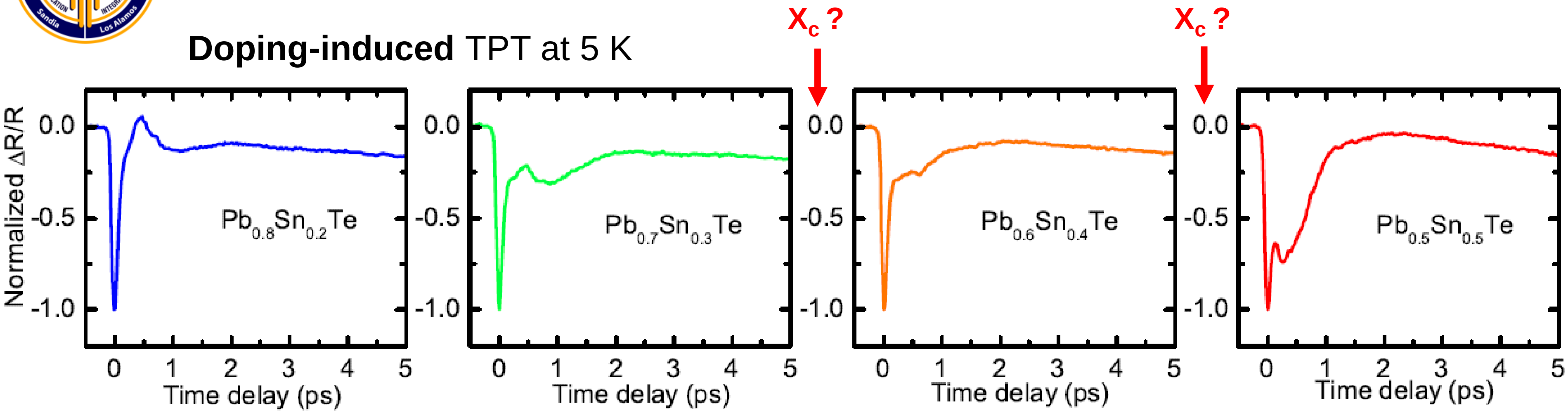
X_c = 0.4 at 5K

Yan et al. PRL **112**, 186801 (2014)

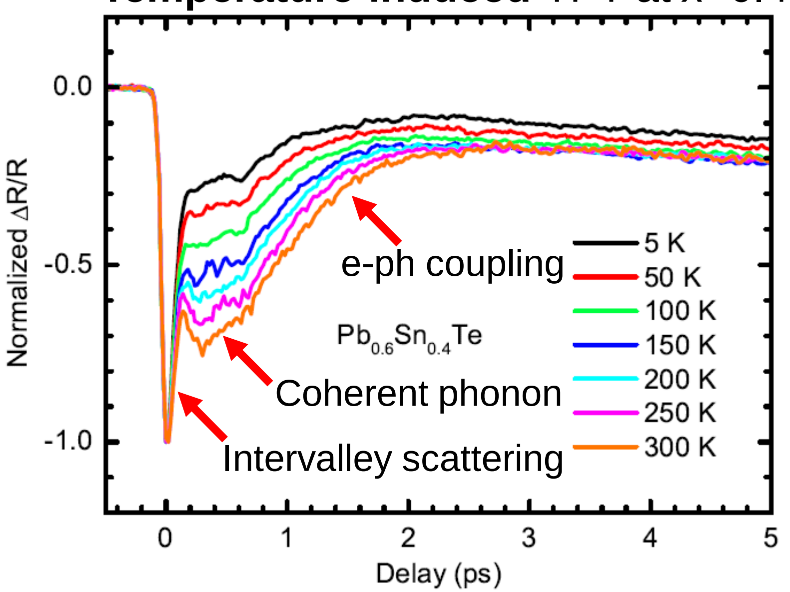
Can we use UOS to find the evidence for TPT with ***temperature*** and ***doping***?

Preliminary Results and Future Directions

Doping-induced TPT at 5 K



Temperature-induced TPT at x=0.4



- Strong electron-phonon coupling in TI state – common to all TI
- Investigate the effect of magnetic field using THz spectroscopy to probe conductivity of photoexcited carriers.
- Apply circularly polarized pump to break TRS and study the dynamics of the ***k***-spin locking process.

Temperature Dependence of Decay Amplitudes